Extinction theorem for ultrafast pulses

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Instead of using a frequency dependent refractive index, we propose to use the extinction theorem to describe reflection and transmission of an ultrashort pulse passing through the boundary. When the duration of the pulse is comparable with the relaxation time, the results differ significantly from those given by the traditional method, especially if the carrier frequency is close to an absorbtion line. We compare the two approaches using the data of GaAs in the infrared domain. ©2000 Optical Society of America

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It is well known that the reflection and refraction of an electromagnetic wave on the boundary of a material system is a light scattering phenomenon. The atoms in the medium are brought into oscillations by the incoming wave, and then secondary waves are generated by the induced atomic dipoles. The transmitted wave is the result of superposition of the scattered wave and the external field, while the reflected wave is a result of backscattering. One side of the process, the solution of the quantum mechanical equations of motion of the perturbed atoms (or the forced oscillations in the classical model) is treated in most textbooks in order to calculate the frequency dependence of the polarizability and the dielectric constant. The other side of the problem, the actual scattering process is usually replaced by phenomenological boundary conditions – Fresnel formulas – for the macroscopic fields. One then uses a frequency dependent index of refraction, and calculates the response for each spectral component.

Instead of that traditional approach which neglects the transient regime of the forced oscillations, we use a procedure that exhibits the actual physical process. This is the method of integral equations, known also as the Ewald-Oseen extinction theorem in the stationary situation.[1] We are going to consider here the one-dimensional variant of this theory applied to ultrafast pulses.

We note that local field effects influence the coupled atom-field dynamics, therefore it can affect the reflection and transmission properties too. For weak fields this leads only to a constant shift[2] of the resonance frequencies, therefore we shall treat the problem here without this correction.

The extinction theorem has been applied for the resonant nonlinear case in our works[2–4] but with the slowly varying amplitude approximation (SVEA) in time. The first model calculation for a linear medium treating the full time development of a pulse without SVEA has been given by one of us.[6]

We consider the boundary value problem for the transmission of a light pulse through a plane-parallel resonant layer consisting of polarizable atoms. Let the incoming field be a linearly polarized plane wave:

$$E_{in}(x,t) = E(t - x/c) \tag{1}$$

We restrict ourselves here to the case of normal incidence. Then the problem is one dimensional in space, thus the macroscopic field obeys the inhomogeneous wave equation:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2 P}{\partial t^2}. \tag{2}$$

In the situation considered here, P is different from zero in a slab placed between $x = 0$ and $x = L$, and the solution of Eq.(2) has the form:[2]

$$E(x,t) = E_{in}(x,t) - \frac{1}{2\varepsilon_0 c}\int_0^L \frac{\partial P}{\partial t}\,(x', t - |x - x'|/c)dx'. \tag{3}$$

Here E_{in} is the solution of the homogeneous equation corresponding to (2), and it is identified with the incoming wave, while the second term is a scattered wave which is the superposition of outgoing elementary waves originating in different x' planes. Given an incoming wave, the transmitted wave is determined by the whole solution (3) at $x \geq L$, while the reflected wave is described only by the second, integral term at $x \leq 0$.

As we are considering the linear case, the dynamics of the polarization P in the medium can be determined by a first order perturbation theory. Supposing that at the beginning the system is in its ground state, the dipole moment density induced by the field is given by the following expression:

$$P(t,x) = \sum_j P_j = 2NRe\left[\sum_j \frac{i|d_j|^2}{\hbar} e^{(-i\omega_j - \frac{\gamma_j}{2})t} \times \int_0^t E(t',x)e^{(i\omega_j + \frac{\gamma_j}{2})t'}dt'\right] \tag{4}$$

where N is the number density of the dipoles in the medium, d_j is the transition dipole matrix element between the ground state and the j-th excited level, ω_j and γ_j are the corresponding transition frequencies and decay constants, and $E(t,x)$ is the field strength at the position x of the atomic dipole. In the ordinary theory of dispersion the time dependence of the electric field, $E(t)$ is taken to be $E_0 e^{-i\omega t}$, and after performing the integration in (4) the terms containing $e^{-\gamma t/2}$ are omitted, as they are rapidly decaying. In this way one obtains a frequency dependent susceptibility $\chi(\omega) = (1/\varepsilon_0)\sum_j (2N|d_j|^2/\hbar)/(\omega_j - \omega - i\gamma_j/2)$, and refractive index $n = (1+\chi(\omega))^{1/2}$. Our main point is that if the duration of the whole pulse is shorter than the decay time of the terms which contain $e^{-\gamma_j t/2}$, this traditional approach fails, and all the terms following from (4) should be kept.

In the case of an optically thin layer, the calculations can be simplified. Then $L/\lambda \ll 1$, the spatial variation of the polarization can be neglected, and instead of integrating in Eq.(3) we can use the mean value of the time derivative of the polarization. In this thin medium case one can derive a system of coupled differential equations for the polarization components, P_j :

$$\ddot{P}_j + \dot{P}_j\,\gamma_j + P_j(\frac{\gamma_j^2}{4} + \omega_j^2) \;=\; 2N\frac{|d_j|^2}{\hbar}(\omega_j E_{in} - \frac{L}{2\varepsilon_0 c}\omega_j \sum_k \dot{P}_k) \qquad (5)$$

For an arbitrary thick layer one has to solve the coupled integro-differential equations (3) and (4), with a numerical method.

We have performed calculations, based on the theory given here and compared the results with those of the traditional method of Fresnel formulas. For definitness we have used the data[5] of GaAs in the infrared domain. Two resonant frequencies and the with constants $\lambda_1 = 37.31\mu m$, $\lambda_2 = 0.4305\mu m$, $\gamma_1 = 0.034\omega_1$, $\gamma_2 = 0$, $(\lambda_i = 2\pi c/\omega_i)$, and $s_i := \dfrac{2\ N|d_i|^2}{\varepsilon_0\hbar\omega_i}$, with $s_1 = 9.89$, $s_2 = 2.07$. reproduced very well the measured[5] stationary dispersion and absorbtion of the material in the range $\lambda = 1 - 10^2\mu m$. For the sake of simplicity we have chosen the following form of the incoming pulse $E_{in}(t) = \sin^2\Omega t\cos\omega_0 t$, $0 < t < \pi/\Omega$. As we are considering the ultrafast regime we have chosen $\Omega = 0.25\omega_0$. The shape of this incoming pulse is shown in Fig. 1(a) below. The solution of Eq.(5) corresponding to this incoming pulse and to the initial conditions $P_j(0) = 0$, $\dot{P}_j(0) = 0$, $j = 1, 2$ can be given for the thin layer by an exact but lengthy formula.

Far from resonance ($\omega_0 \ll \omega_1$, $\omega_0 \gg \omega_1$) both methods predict the same results. On the other hand significant differences can be seen for pulses with $\omega_0 \approx \omega_1$, i.e. close to resonance, but still in the linear regime. The calculations were performed with $\omega_0 = 1.03\omega_1$, for layers of different thicknesses of the order of the central wavelength, $\lambda_0 = 2\pi c/\omega_0$. Fig. 1 shows the results obtained for an optically thin layer $L = \lambda_0/256$. In that case we have solved the system of equations (5). The numerical solution of Eqs. (3) and (4) are shown in Fig. 2 for a layer of thickness of λ_0. The continuous lines are the results obtained from the present transient extinction theory, while the dashed lines are the amplitudes obtained by using the frequency dependent index of refraction. It can be seen, that some of the overall characteristics of the response obtained by the two different methods are similar. Both treatments predict significant broadening of the reflected and transmitted pulses, since the decay time, $2/\gamma$ is long compared with the duration of the excitation. In addition, till the end of the exciting pulse, $\omega_0 t = 4\pi$, the reflected amplitudes are very similar except for a little phase shift. The transmitted pulse appears in both cases with the same expected delay that can be calculated as $t_g = v_g/L$, where v_g is the the group velocity.[7,8] This gives $\omega_0 t_g = 7.96\pi$ in good agreement with the numerical result. We note, in addition, that both methods yielded precursors[7,8] in the transmitted fields appearing at $t = L/c$, not seen in the figures, because of their smallness.

In the case of the thin layer the forms of the fields are quite similar, see Fig 1. On the other hand significant differences are seen in the time dependences of the transmitted and reflected pulses calculated by the two different treatments in layers of thickness comparable with λ_0. We attribute these differences in the amplitudes, as well as in the phases to the approximative character of the time dependence of the polarization in the Fourier method, i.e. to the omission of the transients at the beginning and at the end of the excitation. This part is non-negligible if the duration of the process is comparable with the relaxation time since it represents an important contribution to the elementary waves generated by the induced dipoles.

In conclusion, we find that for ultrafast pulses the properties of a dispersive medium must be calculated by taking care of the transient response of the system. This can be done best by using a fully time dependent treatment as proposed in the present work.

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Fig. 1. Incoming (a), reflected (b) and transmitted (c) amplitudes for a layer of thickness $L = \lambda/256$. In (b) and (c) continuous lines are calculated from the extinction theorem, while he dashed lines are obtained using the index of refraction.

Fig. 2. Reflected (a) and transmitted (b) pulses calculated by the two different methods (see caption of Fig 1.) for the incident pulse shown in Fig. 1 (a), and for a layer of thickness $L = \lambda_0$, In (c) the origin has been shifted to $\omega_0 t = 2\pi$ corresponding to $t = L/c$ (the transit time in vacuum)

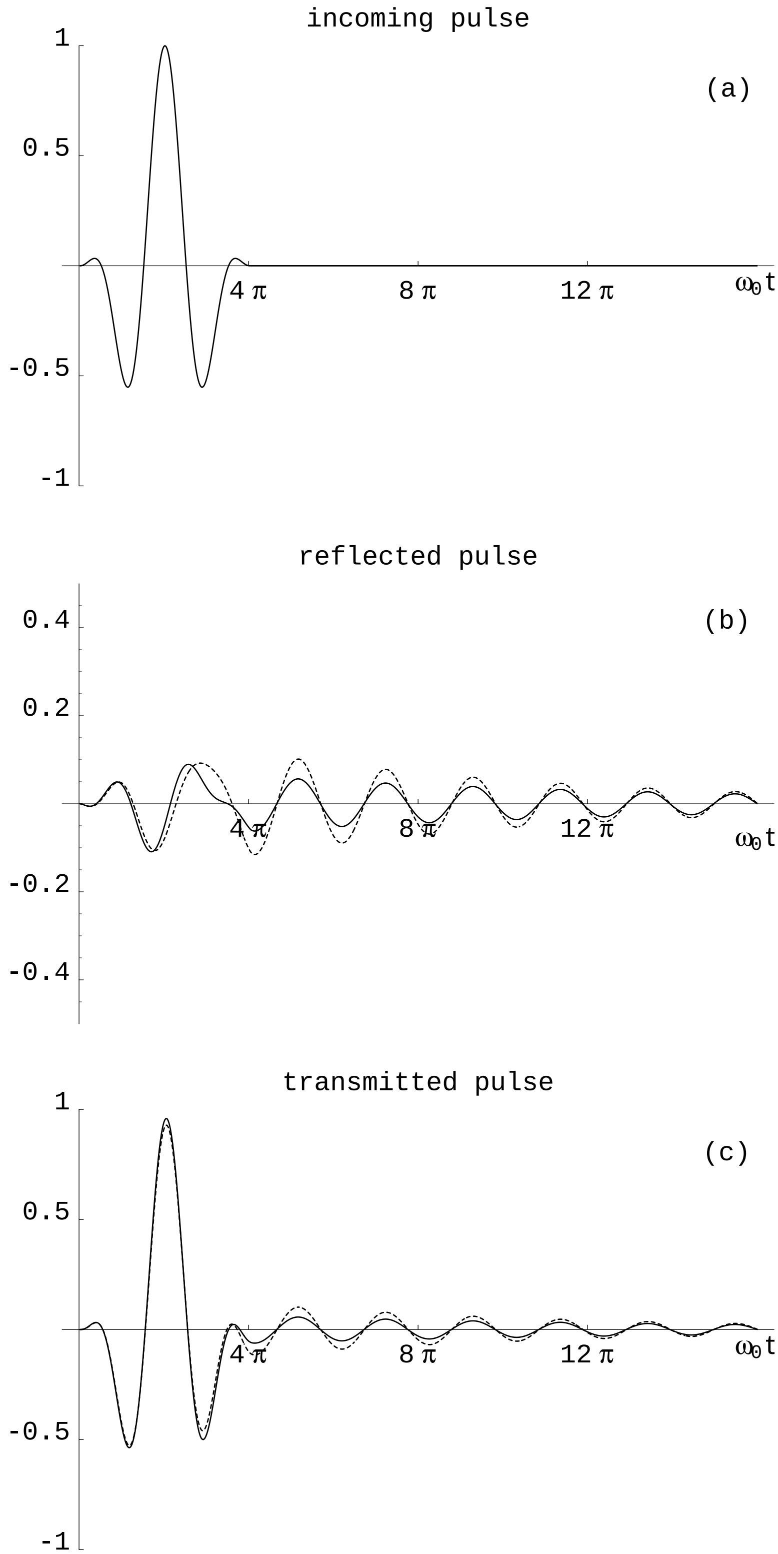
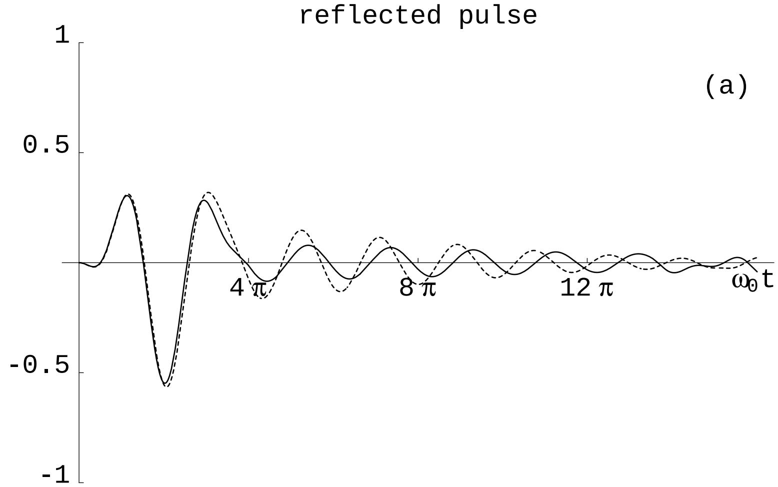


Figure 1. Cs. Benedek, Optics Letters



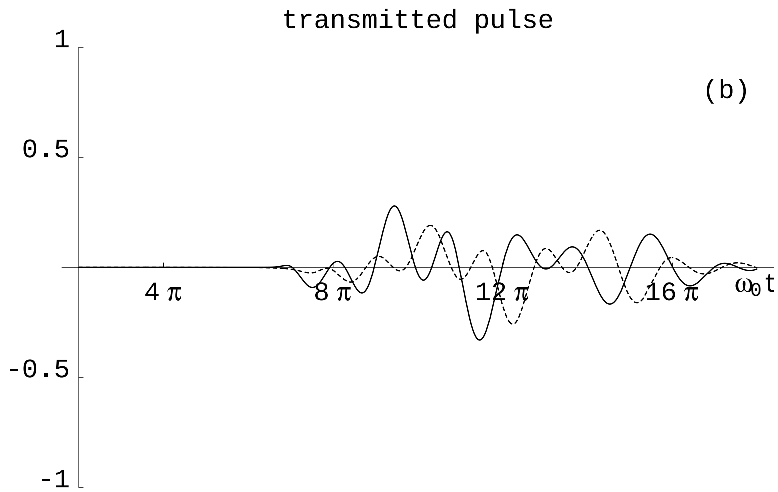


Figure 2. Cs. Benedek, Optics Letters